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Control of Light-Emitting Polymer Devices Using Polymer/Polymer
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13. ABSTRACT (Maximum 200 words)

We present the use of polymer/polymer interfaces to control light-emitting polymer devices. Bilayer devices utilizing poly (9-vinyl carbazole) (PVK) as a hole transporting/electron blocking polymer together with a pyridine containing electron transporting layer show dramatically improved efficiency and brightness as compared to single layer devices. This is attributed to charge confinement and exciplex emission at the PVK/emitting polymer interface. The introduction of emeraldine base (EB) form of polyaniline (PAN) on both sides of the emitting layer enables the device to work under both forward and reverse bias, as well as in AC modes. Interfaces play an important role in the operation of these devices. Furthermore, when the EB is replaced by sulfonated polyaniline (SPAN) on the cathode side and the emitting layer is properly modified to balance electron and hole transport, the device generates different colors of light, red under forward bias and green under reverse bias.

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CONTROL OF LIGHT-EMITTING POLYMER DEVICES USING POLYMER/POLYMER INTERFACES

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ABSTRACT

We present the use of polymer/polymer interfaces to control light-emitting polymer devices. Bilayer devices utilizing poly(9-vinyl carbazole) (PVK) as a hole transporting/electron blocking polymer together with a pyridine containing electron transporting layer show dramatically improved efficiency and brightness as compared to single layer devices. This is attributed to charge confinement and exciplex emission at the PVK/emitting polymer interface. The introduction of emeraldine base (EB) form of polyaniline (PAN) on both side of the emitting layer enables the device to work under both forward and reverse bias, as well as in AC modes. Interfaces play an important role in the operation of these devices. Furthermore, when the EB is replaced by sulfonated polyaniline (SPAN) on the cathode side and the emitting layer is properly modified to balance electron and hole transport, the device generates different colors of light, red under forward bias and green under reverse bias.

INTRODUCTION

Electroluminescence (EL) combined with other unique properties of polymers, such as solution processibility, band gap tunability and mechanical flexibility, make conjugated polymers excellent candidates for low cost large area display applications [1-4]. Most "conventional" polymer light-emitting devices have been shown to be tunneling diodes and can be operated only under forward DC bias [5]. Among the most important limitations associated with many of the "conventional" polymer light-emitting diodes (LEDs) are poor stability and shelf lifetime. The double charge injection mechanism of the "conventional" polymer LEDs requires the matching of the cathode (anode) work function to the corresponding LUMO (HOMO) level of the polymer in order to achieve efficient charge injection. The relatively low electron affinity of most conjugated polymers requires metals with very low work functions to achieve efficient electron injection.

Pyridine-containing conjugated polymers are promising candidates for light-emitting devices [6]. The pyridine containing polymers are highly luminescent, especially the copolymers. The high electron affinity of pyridine based polymers enables the use of relatively stable metals such as Al, Au, or ITO and doped polyaniline as electrodes. Taking advantages of the better electron transport properties of the pyridine-containing polymers, we fabricated bilayer devices utilizing poly(9-vinyl carbazole) (PVK) as hole transporting/electron blocking polymer, which improves the device efficiency and brightness significantly due to the charge confinement and exciplex emission at the PVK/emitting polymer interface [7]. By inserting a layer of emeraldine base (EB) form of polyaniline on both side of the emitting polymer, we fabricated symmetrically configured AC light-emitting (SCALE) devices [8] which work under both forward and reverse DC bias as well as in AC modes. When we replace the EB layer on the cathode side of the SCALE device with sulfonated polyaniline (SPAN) and modify the emitting layer to appropriately balance

electron and hole transport, color variable bipolar/AC light-emitting devices are fabricated that can generate different colors of light depending on the polarity of the driving voltage, red under forward bias and green under reverse bias [9]. Figure 1 shows the repeat units of the pyridine-containing polymers and other polymers that were used in these devices.

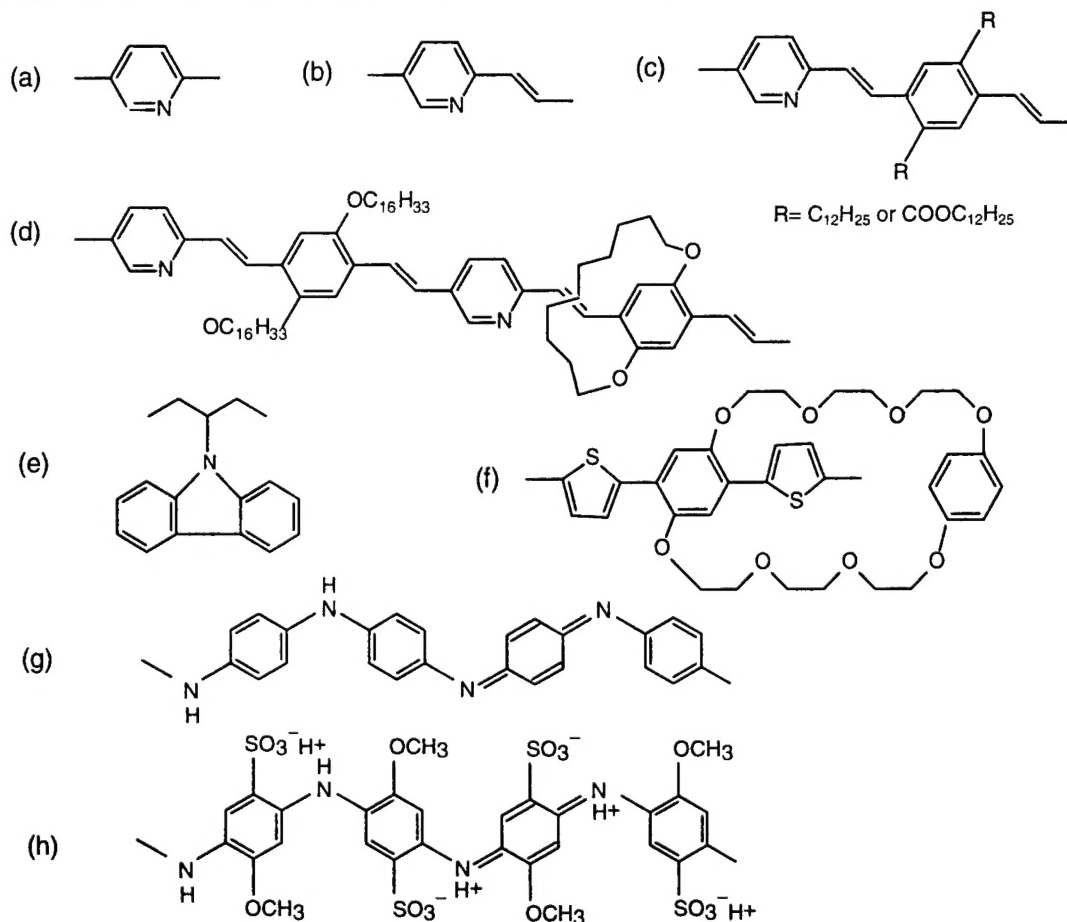


Fig. 1 Repeat units of the pyridine-containing polymers and other polymers used in the study. (a) poly(p-pyridine) (PPy); (b) poly(p-pyridyl vinylene) (PPPyV); (c) copolymer of PPV and PPyV (PPyVP(R)₂V); (d) wrapped copolymer of pyridyl vinylene and phenylene vinylene (@PPyVPV); (e) poly(9-vinyl carbazole) (PVK); (f) wrapped copolymer of dithienylene and phenylene (@PTP); (g) emeraldine base (EB) form of polyaniline; (h) sulfonated polyaniline (SPAN).

EXPERIMENT

For bilayer devices, the PVK layer was spin coated onto the ITO substrate from solution in tetrahydrofuran (THF) (~10 mg/ml) at ~3000 rpm. The emitting layer was then spin coated on top of the PVK layer from appropriate solutions (typically xylenes). As the solvent for the emitting layer does not dissolve PVK, no significant intermixing of the two polymers is expected. The SCALE devices were fabricated by spin-casting a solution of EB in N-methyl pyrrolidinone (NMP) (~5 mg/ml) onto an ITO substrate. After drying in dynamic vacuum, a layer of the emitting polymer was spin-coated on the EB surface. Another EB layer was similarly coated on top of the emitting layer. The spinning speed for all layers is ~ 2000 rpm. The color

variable/AC light-emitting devices were similarly fabricated as the SCALE devices except that the top EB layer was replaced by SPAN cast from aqueous solution and the emitting layer was comprised of a blend of polymers. All the spin coating procedures were carried out inside a class 100 cleanroom in air. The top metal electrodes were deposited by vacuum evaporation at a pressure below 10^{-6} torr. To prevent damage to the polymers, the substrates were mounted on a cold-water cooled surface during the evaporation.

Absorption spectra were measured on spin-cast films using a Perkin-Elmer Lambda 19 UV/VIS/NIR spectrometer. Photoluminescence (PL) and EL were measured using a PTI fluorometer (model QM-1). The current-voltage characteristics were measured simultaneously with EL using two Keithley model 195A multimeters while dc voltage was applied by a HP model 6218A DC power supply. We note that all the device testing procedures were performed in air on as-made devices without any encapsulation.

RESULTS AND DISCUSSION

The performance of a bilayer devices using PVK as the hole transporting/electron blocking layer improves dramatically as compared to single layer devices. The internal quantum efficiency and brightness of the bilayer devices increase two to three orders of magnitude, reaching $\sim 0.5\%$ and over 300 cd/m^2 , respectively. Figure 2 compares the electroluminescence-voltage and electroluminescence-current characteristics for a single layer device and a bilayer device using the wrapped copolymer as the emitting layer. PVK enhances the transport of holes injected from the anode and blocks the transport of electrons injected from the cathode such that the electrons accumulate at the PVK/copolymer interface. In addition, the PVK layer removes the recombination zone from the vicinity of the electrode so that the radiative recombination is protected against the non-radiative quenching at the electrode/polymer interfaces.

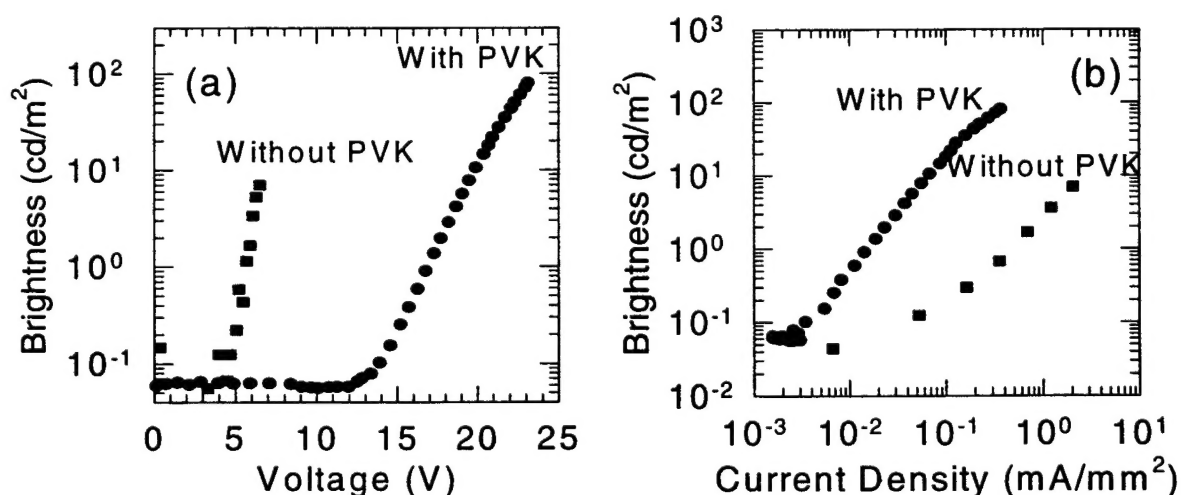


Fig. 2 Comparison of (a) brightness-voltage and (b) brightness-current characteristics for a single layer device (square) and a bilayer device (circle).

Quenching at the electrodes has been reported for phenylene vinylene oligomers by Park *et al.* [10] and in MEH-CN-PPV by Becker *et al.* [11]. Becker *et al.* also note that the removal of the recombination zone away from the metal electrode to a polymer/polymer interface is crucial to increasing the EL efficiency in their PPV/MEH-CN-PPV bilayer devices [11]. In order to probe

the effect of the electrodes on the luminescence properties of our single and bilayer films a number of samples were fabricated on ITO. Electrodes were evaporated on single layers of PVK and wrapped copolymer @PPyVPV. On each film the PL was measured in the vicinity of and far from the electrode with the exciting light entering the sample from the ITO side and recording the EL from the ITO side. The insets of Fig. 3 show the device configurations. The relative PL intensities of PVK at these two locations for 330 nm excitation wavelength (Fig. 3a) shows that the PL increases modestly when the excitation light is incident on the electrode. In the absence of quenching an increase in PL intensity is expected for two reasons: reflection of the excitation light by the Al and reflection of the photoluminescence by the Al. The small increase for PVK indicates significant quenching near the Al electrode. Figure 3b shows the same experiment for a single layer of wrapped copolymer @PPyVPV using a 450 nm wavelength excitation. The nearly two-fold decrease in PL intensity indicates extensive quenching by the Al electrode for this polymer.

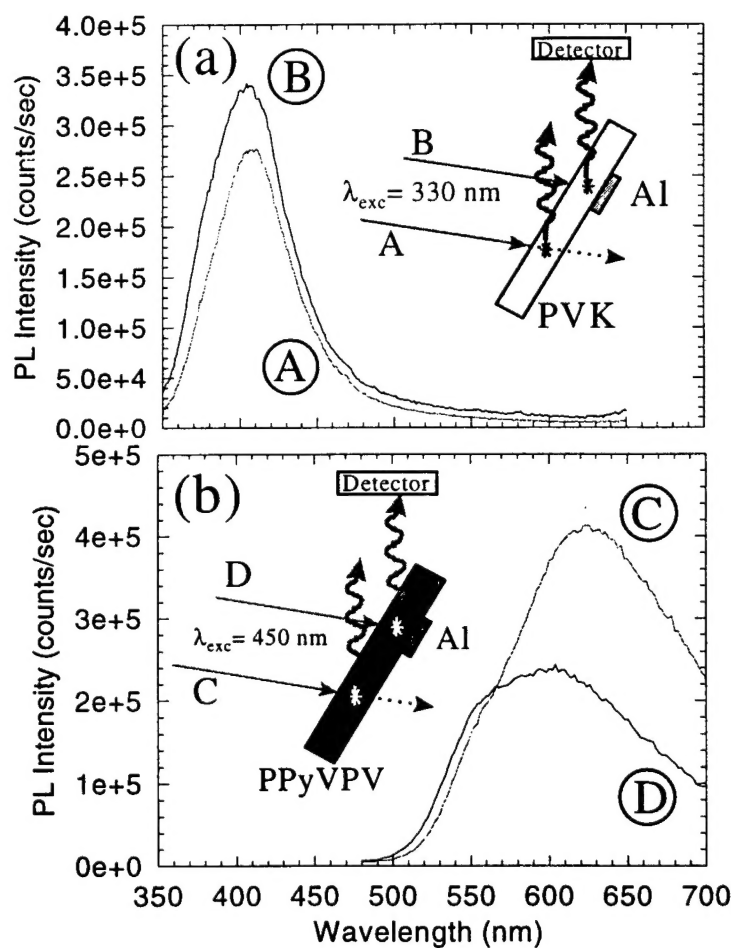


Fig. 3 PL of films of PVK (A,B) and PPyVPV (C,D) for the configurations shown schematically in the insets. (a) 330 nm excitation and (b) 450 nm excitation.

For bilayers in which the luminescence occurs primarily at the interface the PL and EL intensity should be unaffected by the presence of the electrode, i.e., no quenching should occur. Figure 4b shows the PL intensity for a PVK/@PPyVPV bilayer with a 330 nm excitation (experimental configuration shown in Fig. 4a). When the excitation wavelength is incident on the evaporated Al electrode the PL intensity increases dramatically. The increase is attributed to the reflection of the luminescence and the excitation light implying that the emission is primarily

occurring at the polymer/polymer interface. Figure 4c shows the PL intensity of the bilayer when the lower energy excitation is used (450 nm). In this case the PL intensity is also increased, but the emission is shifted away from the single layer emission to the exciplex (interface) emission, when the exciting light is incident on the reflecting electrode. Although the low energy excitation is used, the emission at the interface is observed because the wrapped copolymer @PPyVPV PL is quenched at the electrode and the interface emission is enhanced. Energy is transferred from excitons in the wrapped copolymer @PPyVPV to exciplexes at the interface. The diffusion coefficient for excitons in the @PPyVPV copolymer can be estimated from the diffusion distance $\sim 10^{-6}$ cm) and the lifetime of the exciton in the copolymer (~ 1 ns) [12], giving a diffusion coefficient of $\sim 10^{-3}$ cm²/s. Thus, exciplex emission in the bilayer devices substantially reduces the effects of quenching by electrodes in single layer devices.

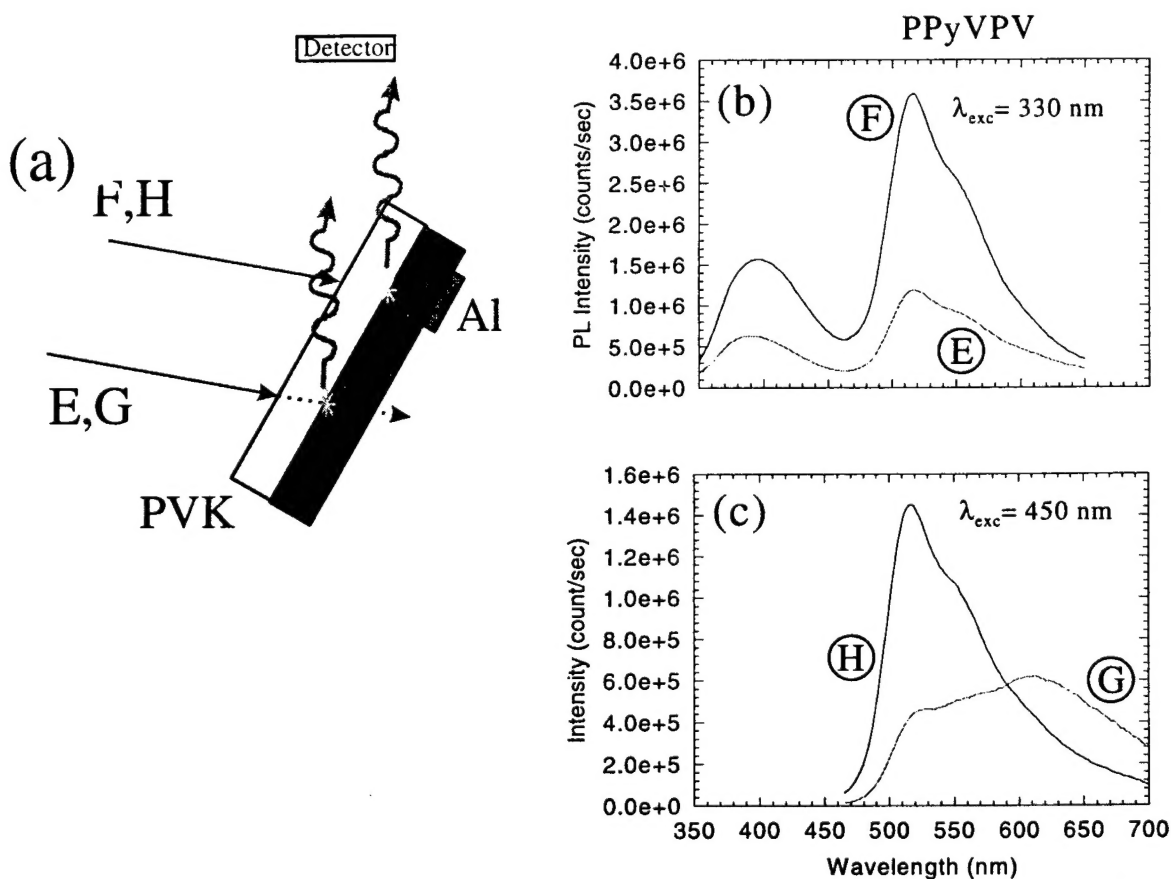


Fig. 4 PL of bilayer films of PVK/PPyVPV with the configuration shown schematically in (a). The excitation energies were (b) 330 nm (E,F) and (c) 450 nm (G,H).

An unwanted side effect of using the PVK layer is that it increases the device operating voltage substantially. One effective way to reduce the device turn on voltage is to use a high surface area network electrode [13]. The rough electrode will create a non-uniform high electric field that enhances the charge injection. This technique has been successfully applied to PPV based single layer devices [14].

The light generation in the bilayer device is attributed to decay of exciplexes formed at the PVK/copolymer interface. Figure 5 compares the PL of pure wrapped copolymer, pure PVK, and bilayer of PVK/copolymer, as well as the EL spectra of the bilayer device. The PL of PVK film excited at 3.6 eV has an emission peak at 3.06 eV. The PL of pure wrapped copolymer film excited at 2.65 eV peaks at 2.03 eV with a shoulder at 2.25 eV. When the PVK/copolymer bilayer film is excited at 3.6 eV, a new peak appears at 2.38 eV with another peak at 2.23 eV. The new peak is assigned to exciplex emission at the PVK/copolymer interface, supported by the optical absorption and photoluminescence excitation (PLE) measurements. The EL of the bilayer device follows closely with the PL of the bilayer film in accord with the EL emission originating primarily from the exciplex formed at the PVK/copolymer interface.

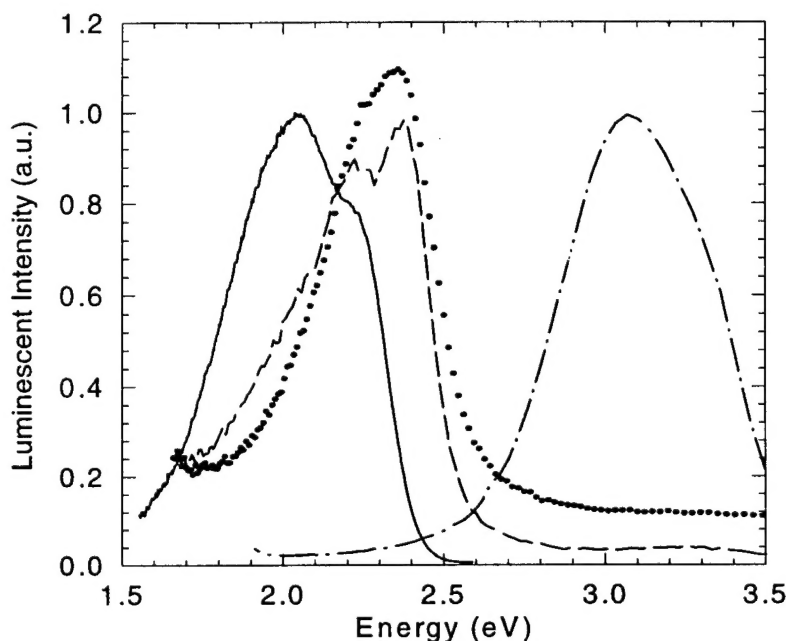


Fig. 5 Normalized PL spectra of pure wrapped copolymer film (solid line) excited at 2.65 eV, pure PVK film (dash-dotted line) excited at 3.6 eV, bilayer of PVK/copolymer (dashed line) excited at 3.6 eV, and the EL of the bilayer device (filled circle). The EL spectra are offset for clarity.

Two novel device configurations that can be operated in both forward and reverse DC bias as well as in AC modes are the SCALE devices and color variable bipolar/AC light-emitting devices. The SCALE devices consist of a light-emitting layer sandwiched between two "insulating" redox polymer layers, then placed between two electrodes. The SCALE device configuration is quite general, it can be applied to a variety of emitting and redox polymers, as well as electrodes. For the SCALE device presented here, PPy was used as the emissive layer; the emeraldine base (EB) form of polyaniline was utilized as the redox material; ITO and Al were used as electrodes. Figure 6 inset shows schematically the structure of such SCALE devices. They emit light under both forward and reverse DC bias as well as under AC driving voltage. Under low frequency AC (sinusoidal) driving voltage, nearly uniform light pulses with twice the driving frequency were observed, Figure 6. This unusual behavior is attributed to the effects of charge accumulation at the polymer/polymer interfaces [8]. We propose the following mechanism for the SCALE device operation. Under low bias voltages, electrons and holes can be injected from the electrodes into the quinoid and benzenoid levels of EB and form negative and

positive polarons, respectively. These polarons transport to the EB/PPy interfaces via a hopping mechanism and populate the EB/PPy interfaces at the polaron levels. Because the polarons levels are within the π - π^* band gap of EB, and are also likely within the band gap of PPy, the barriers for charge injection from electrodes to polaron levels of EB are significantly reduced as compared to injection directly to conduction and valence bands of PPy. Within this model, the limiting barriers for charge injection are changed from the electrode/polymer contacts as proposed for conventional polymer LEDs to the polymer/polymer (EB/PPy) interfaces of the SCALE devices. When the applied electric field is high enough, the stored charges begin to tunnel into the conduction and valence bands of PPy. When they meet, the injected charges may form intrachain excitons and decay radiatively to emit photons or follow other nonradiative decay paths. If the charge injection is not balanced, as is the case for most polymer LEDs, the excess charge carriers may migrate through the PPy layer without decaying. Most of these charges will be trapped in the opposite PPy/EB interface. When the bias voltage is reversed, the shallow trapped charges will be released from the interfaces and contribute to the recombination current. The deep trapped charges which act as quenchers or injection limiters in DC devices will be neutralized. We point out that the use of stable high work function metals, such as Au, as electrodes to inject both electrons and holes for the SCALE devices may reduce the problems of aging of contacts of polymer light-emitting devices. Also, continuous reversal of the sign of the driving voltage under AC operation may reduce degradation.

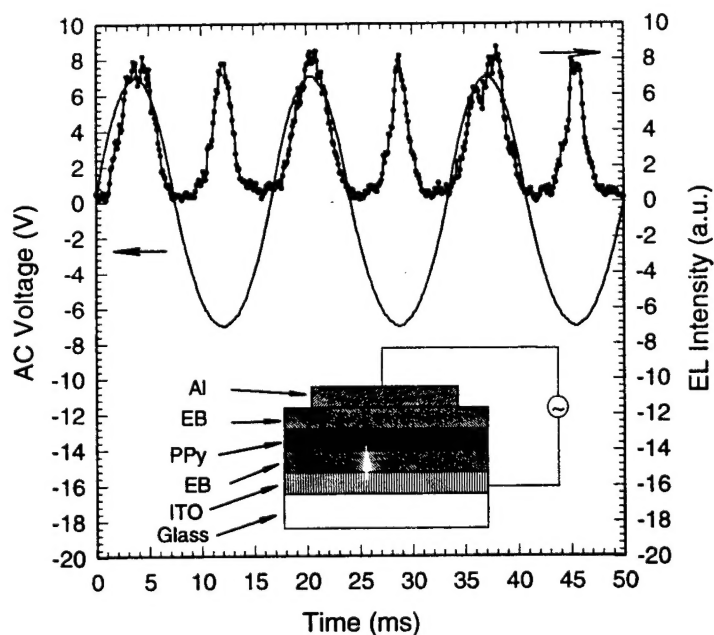


Fig. 6 EL intensity as a function of time for a SCALE device driven by a 60 Hz sinusoidal voltage. Inset: schematic device structure of the SCALE device.

We have reported a new approach to the color variable polymer light-emitting devices based on SCALE device structure: color variable bipolar/AC light-emitting devices [9]. In this approach we replace the EB layer on the cathode side of the SCALE device structure with sulfonated polyaniline (SPAN). Because the SPAN is capable of modifying the emission properties of certain pyridine containing polymers at the interface such that the interface emits

different colors of light than the bulk does, the device is able to generate a different color of light under forward and reverse bias. In this approach, the colors of light are controlled by selecting the desired emission locations which in turn are controlled by the polarity of driving voltage and the charge injection and transport properties of the polymer layers. Since motion of ionic species is not required for device operation a relatively fast time response is achieved, allowing the colors to be switched rapidly.

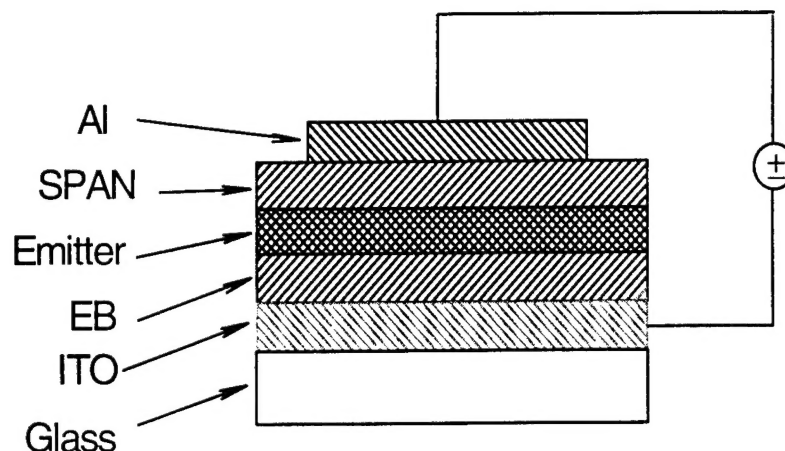


Fig. 7 Schematic structure of the color variable light-emitting devices

For the devices presented here, the wrapped copolymer @PPyVPV, and a wrapped copolymer of polythiophene and polyphenylene derivative, @PTP, were used as the emitting materials; SPAN and EB were used as the redox materials; ITO and Al were used as electrodes. Figure 7 shows the schematic diagram of the device structure of the color variable bipolar/AC light-emitting devices. Figure 8 shows the typical current-voltage and brightness-voltage characteristics of the color variable devices. The devices have typical turn on voltages of ~ 4 - 8 V depending upon film thickness and work equally well under both polarities of driving voltage with different colors of light being emitted. The light appeared red and green to the eye under forward and reverse bias, respectively, and was clearly visible under normal indoor lighting. Internal device efficiencies of 0.1% photons/electron has been achieved for the initial devices. Figure 9 shows the EL spectra of the color variable device under forward and reverse bias. For comparison, the EL spectra for single layer @PPyVPV and @PTP are also shown. The CIE chromaticity x,y coordinates of the two spectra are calculated to be (0.654,0.345) and (0.471,0.519), respectively, showing both colors to be relatively pure (see Fig. 8 inset). The colors of the devices can be switched rapidly, up to ~ 20 kHz, depending upon device impedance and geometry.

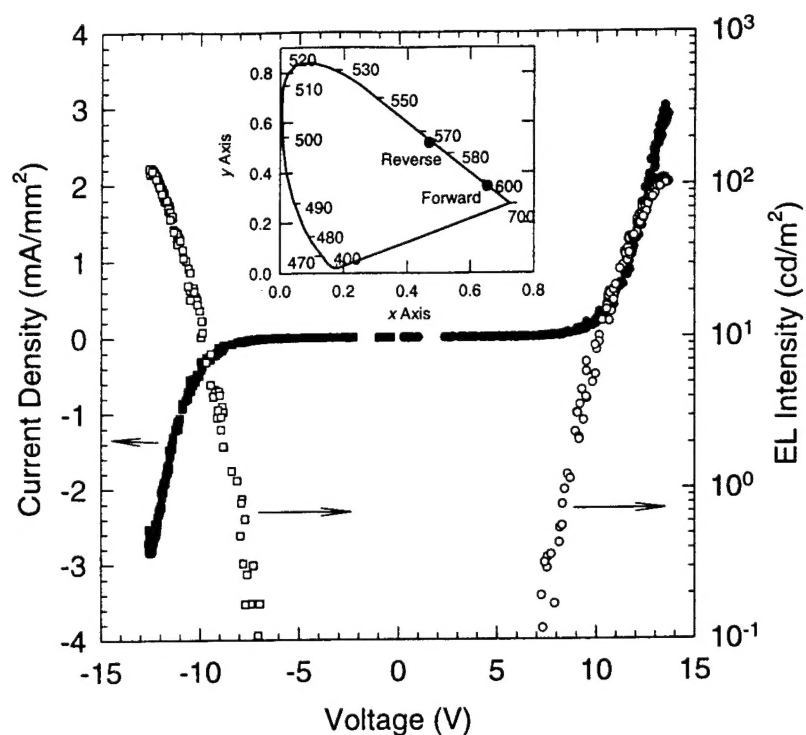


Fig. 8 Current-voltage and brightness-voltage characteristics of a color variable light-emitting device under forward and reverse bias conditions. Inset shows the colors of the device under forward and reverse bias in the CIE chromaticity diagram.

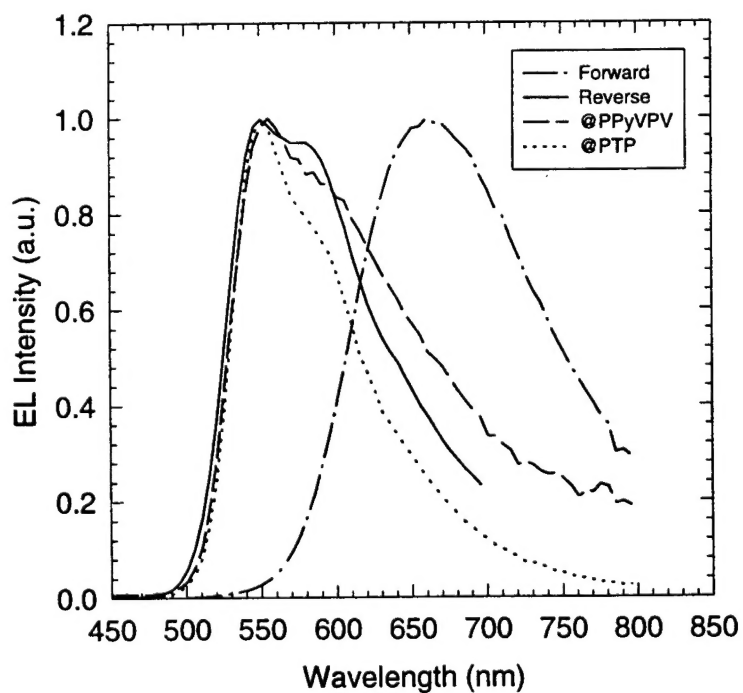


Fig. 9 EL spectra of the color variable device under forward and reverse bias conditions. For comparison, the EL spectra for single layer PPyVPV and PTP devices also are shown.

The EL spectrum under forward bias is substantially different from that of the single layer devices of either @PPyVPV or @PTP, suggesting that the light is generated from the interface between the emitter blend and either EB or SPAN under forward bias. To further clarify this, we fabricated the following devices: ITO/@PPyVPV/Al; ITO/SPAN/@PPyVPV/Al; ITO/@PPyVPV/SPAN/Al; ITO/SPAN/@PPyVPV/SPAN/Al and similar devices replacing SPAN with EB. Only ITO/@PPyVPV/SPAN/Al and ITO/SPAN/@PPyVPV/SPAN/Al show dramatically redshifted EL. Similar studies using @PTP as the emitting layer show that the emission properties of @PTP are not affected significantly by the presence of the SPAN layer.

The EL spectrum of the color variable device under reverse bias are similar to those of the single layer @PPyVPV and @PTP devices implying that the light is generated either in the bulk of the emitting polymer or at the EB interface. The EB layer functions as a charge injection enhancement layer, playing a similar role as it does in SCALE devices reported earlier [8]. Under reverse bias, the SPAN layer on the cathode side plays a similar role.

It is noted that when the blend in the color variable devices is replaced by pure @PPyVPV polymers, the devices emit red light in forward bias and red-orange light in reverse bias. This indicates that under reverse bias the light is still generated near @PPyVPV/SPAN interfaces. This can be understood in terms of different electron and hole transport properties of the @PPyVPV and @PTP polymers. It is known that most conjugated polymers, including poly(phenylene vinylene) (PPV), poly(*p*-phenylene) (PPP) and polythiophene, have better hole than electron transport properties [3]. The addition of a high electronegativity unit, pyridine, to the backbone is expected to improve the electron transport properties. Therefore, under reverse bias the light is still generated near the SPAN interface for the ITO/EB/@PPyVPV/SPAN/Al device. By adding @PTP, which is expected to have better hole transport properties, to @PPyVPV, the overall hole transport properties of the blend is improved, and hence the light is generated away from the SPAN interface for the ITO/EB/Blend/SPAN/Al device under reverse bias, emitting green light. The mechanism for the SPAN layer changing the emission properties of the @PPyVPV polymer is attributed to the formation of new emissive species due to protonation of the pyridyl units by SPAN. The new species are emissive and emit redshifted light as compared to the non-protonated polymer.

SUMMARY AND CONCLUSION

In summary, we have presented a number of DC and AC light-emitting devices based on the role of polymer/polymer interfaces. The high electron affinity of pyridine based polymers enables the use of relatively stable metals such as Al as efficient electron injecting contacts. Taking advantages of the better electron transport properties of the pyridine-containing polymers, we have fabricated bilayer devices utilizing PVK as hole transporting/electron blocking polymer. The bilayer device structure improves the device quantum efficiency and brightness significantly due to the charge confinement and exciplex emission at the PVK/emitting polymer interface. The fabrication of novel devices such as SCALE devices and color variable bipolar/AC light emitting devices opens new possibilities for device design and operation.

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